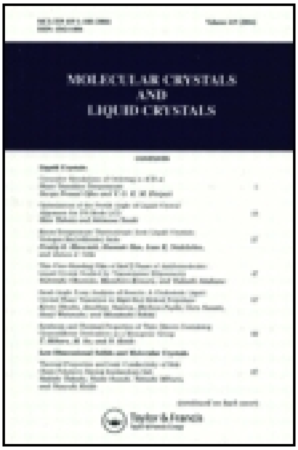

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Flow Properties of Nematic 8CB: An Example of Diverging and Vanishing α_3

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Flow Properties of Nematic 8CB: An Example of Diverging and Vanishing α_3

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The temperature dependence of the Leslie viscosities α_2 and α_3 has been measured in the nematic liquid crystal 8CB (octyl-cyano-biphenyl), using a torsional shear flow apparatus. In most nematics studied so far α_3 is found to be everywhere negative, but if the nematic has an adjacent smectic A phase at lower temperatures, it can be expected that α_3 changes sign and becomes positive in the vicinity of T_{NA} as a precursory phenomenon of the transition. In fact this influence is dominating the major part of the nematic phase.

We have found that for 8CB $\alpha_3 > 0$ in the whole nematic range (33.5°C–40.1°C), except for a small temperature interval of one degree just below the clearing point. This leads to intrinsic flow instabilities, but the flow can be stabilized by applied electric fields. The results further contain the first measurement of a diverging α_3 in a flow experiment.

1 INTRODUCTION

The orientational effects when introducing a shear flow in a nematic liquid crystal are known to belong to one of two categories, which are determined by the sign of two of the five independent Leslie viscosities. These two are commonly denoted α_2 and α_3. If we further assume that $\alpha_2 < 0$ (which has always been found to be the case), it is the sign of α_3 which will be the determining factor for the behaviour of the flow.

If $\alpha_3 < 0$ there will be a stable situation and for high enough shear rates there will be "flow alignment" of the director.[1] This situation is pictured in

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Figure 1, where we also introduce our coordinates and their reference directions. The choice of the angle θ (in favour of $90 - \theta$, which is more commonly used in the literature) is more convenient for homeotropic conditions. For low shear rates the elastic torque Γ_y^E will be important throughout the whole sample and we will get a configuration as in Figure 1a. For high shear rates Γ_y^E will be unimportant compared to hydrodynamic torques, except at two thin boundary layers of thickness ξ near the plates, and the flow will determine the tilt (the flow alignment angle is denoted θ_n). Here we idealize the director configuration as in Figure 1b. The relative importance of elastic and hydrodynamic effects is given by the Ericksen number[2] Er. If Er is sufficiently large ($> 10^2$) elastic effects can be neglected. In the present work this is the case.

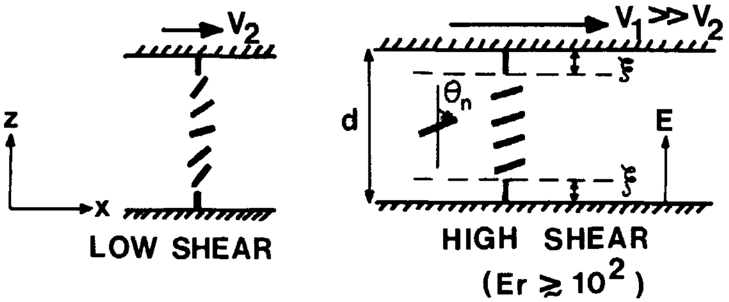


FIGURE 1 a) At low shear the boundary conditions are important throughout the whole sample. b) The picture shows the idealized director configuration for large values of the Ericksen number. At high shear the effects of the boundary conditions on the director are limited to two layers of thickness ξ, and in the bulk of the sample the director makes a constant angle θ_n to the normal. E is the applied electric field $E = \sqrt{2}E_0 \cos 2\pi\nu t$.

The possibility that $\alpha_3 > 0$ has recently become a subject of great interest and experimental evidence has been given that this situation really occurs in the nematic liquid crystals HBAB (hexylo-amino-benzo-nitrile) and CBOOA (cyano-benzylidine-octyloxy-aniline).[3-5] We have earlier reported[6] that this is also the case in the nematic liquid crystal 8CB (octyl-cyano-biphenyl) and in this paper we present measurements of α_2 and α_3 in the nematic range (33.5°C–40.1°C). To understand what happens we refer to Figure 2a, where we have illustrated the hydrodynamic torque Γ_y^h. In our reference frame we have

$$\Gamma_y^h = (|\alpha_2| \cos^2 \theta + \alpha_3 \sin^2 \theta)\frac{dv}{dz} \tag{1}$$

In the absence of external fields, this is also equal to the total torque Γ_y^{tot} which will consequently be positive if $\alpha_3 > 0$. Since the presence of a flow alignment angle requires that Γ_y^{tot} vanishes, such an equilibrium angle will not exist in that case. As indicated in Figure 2b an applied electric field will give an additional (electric) torque Γ_y^{ε} given by

$$\Gamma_y^{\varepsilon} = -\tfrac{1}{2}\varepsilon_a E_0^2 \sin^2 \theta \qquad (2)$$

This torque is negative provided θ lies in the interval $[0, \pi/2]$ ($\varepsilon_a > 0$ in our case) and therefore acts stabilizing on the director. If a strong enough field is used we can regain the stable situation pictured in Figure 1a. This will be used in our experimental determination of α_2 and α_3.

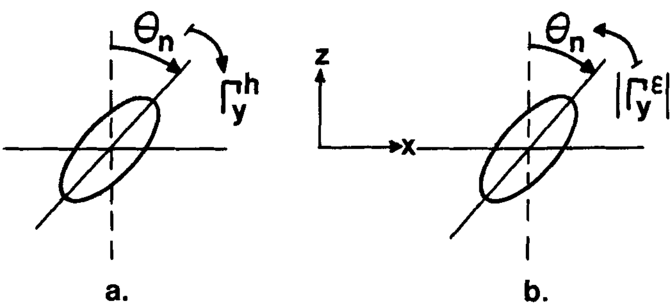


FIGURE 2 a) Direction of the hydrodynamic torque $\Gamma_y^h (\Gamma_y^h > 0$ for $\alpha_3 > 0)$. b) Direction of the electric torque $\Gamma_y^{\varepsilon} (\Gamma_y^{\varepsilon} < 0)$.

II EXPERIMENTAL

In the experiments we use a torsional shear flow apparatus described earlier.[7,8] A schematic picture of the experimental set-up is shown in Figure 3.

The liquid crystal 8CB (purchased from BDH, England) is introduced between two circular glass plates, treated chemically to give homeotropic boundary conditions for the director. When the lower plate is rotated slowly, the liquid crystal is studied between crossed polarizers in parallel light from a HeNe-laser. Due to shear introduced birefringence, increasing radially outwards, an interference pattern of concentric dark rings is seen. This ring pattern contains information about how much the director is distorted in the flow.

The condition for a stationary director in the sample is that the sum of the torques (the total torque Γ_y^{tot}) should vanish. In this case the stationary ring pattern is photographed and analyzed as described in Section IV.

If, on the other hand, the torque is non-zero for shear above a certain limit (outside a certain radius in our shear geometry), there will in this outer region be a markedly different director configuration. The boundary between the "ordered" region (where the director angle $\theta < 90°$) and the outer regions is studied. One advantage of torsional shear compared to linear shear (the possibility to study the flow for long times) is of importance here, since the relevant relaxation times are very long (of the order of minutes to hours).

III THEORY

As mentioned earlier there can be no flow alignment in the shear flow when $\alpha_3 > 0$. To restore the stationary state an electric field is applied across the sample. We will show in detail elsewhere,[9] that shear flow with $\alpha_3 > 0$ and with a strong enough applied electric field in much resembles the situation when $\alpha_3 < 0$ and without an electric field applied. We have derived earlier[7] an expression for the flow alignment angle of a nematic liquid crystal in a shear flow under applied electric field and rewrite Eq. 7 in that work as

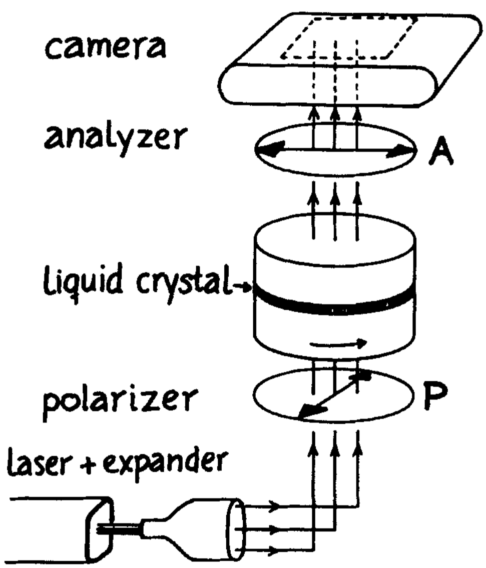


FIGURE 3 Schematic picture of the experimental set-up. The laser light beam is expanded and enters the temperature chamber (not shown), where the liquid crystal flow cell is situated. The interference patterns seen in the flowing liquid crystal are recorded on a photographic film.

follows

$$\tan\theta_n = \frac{\varepsilon_a E_0^2}{2u'^2\alpha_3^2} \genfrac{}{}{0pt}{}{(+)}{-} \left(\frac{\varepsilon_a^2 E_0^4}{4u'^2\alpha_3^2} - \frac{|\alpha_2|}{\alpha_3}\right)^{1/2} \tag{3}$$

When $\alpha_3 > 0$, the minus sign in Eq. 3 has to be chosen by stability conditions.[9] We see immediately that without the electric field the argument in the square root will be negative which implies a non-existing equilibrium angle θ_n. With a non-zero field we can compensate for the negative term in the square root and we see that there will be a lower limit for the stabilizing electric field which we will call the critical electric field, E_c. This field is obtained by setting the argument in the square root equal to zero yielding

$$E_c^2 = \frac{2u'}{\varepsilon_a}|\alpha_2|\alpha_3 \tag{4}$$

The corresponding flow alignment angle θ_c is given by

$$\tan\theta_c = (|\alpha_2|/\alpha_3)^{1/2} \tag{5}$$

Measuring E_c and θ_c would immediately give α_2 and α_3. However, as we can show,[9] the relaxation time τ and the boundary layer thickness ξ will both go to infinity when E_0 equals the critical electric field E_c, invalidating our assumption for the director profile (see Figure 1b). Therefore we instead describe the electric field E_0 by a real number f according to

$$f = \frac{E_0}{E_c} \tag{6}$$

and derive[9] the following expressions for θ_n and ξ as function of f (i.e. E_0)

$$\tan\theta_n = \left(\frac{|\alpha_2|}{\alpha_3}\right)^{1/2} \{f^2 - (f^4 - 1)^{1/2}\} \tag{7}$$

$$\xi = \xi_0(f^4 - 1)^{-1/4} \qquad \xi_0 = \left(\frac{K}{2u'|\alpha_2|\alpha_3}\right)^{1/2} \tag{8}$$

which are valid whenever $f > 1$, i.e. for fields larger than E_c.

IV RESULTS AND DISCUSSION

Measurements were made at nine temperatures in the nematic interval. Different experimental methods were used for positive and negative α_3: For temperatures where $\alpha_3 > 0$ (below 39.1°C) an a.c. electric field (frequency 500 Hz) was applied, and the radius of the stability–instability boundary was measured after the torsional shear had lasted for 90 minutes. A photograph of this situation is shown in Figure 4a. With the critical electric field E_c and the velocity gradient u' we get the product $\alpha_2\alpha_3$ from Eq. 4. The required values for the dielectric anisotropy ε_a were taken from Ref. 10. To separately determine α_2 and α_3 we measure the angle θ_n for $f = 1.5$. This is achieved by making the experiments at two different sample thicknesses ($d = 600\,\mu$m and $300\,\mu$m) as described in Ref. 7. Equation 7 then gives the angle θ_n and the ratio α_2/α_3. We have used values for the refractive indices from Ref. 11. An example of an interference pattern is seen in Figure 4b.

For $\alpha_3 < 0$ we determine α_2 and α_3 by studying the influence of a weak electric field on the flow alignment angle. This directly gives α_2 and α_3 through Eq. 15 of Ref. 7:

$$\tan\theta_n = \left(\frac{\alpha_2}{\alpha_3}\right)^{1/2} + \frac{\varepsilon_a E_0^2}{(2u'\alpha_3)} \tag{9}$$

The measured behaviour of $\alpha_2(T)$ and $\gamma_1(T)$ ($\gamma_1 = \alpha_3 - \alpha_2$) is shown in Figure 5, and the corresponding behaviour of $\alpha_3(T)$ in Figure 6. The Leslie viscosity α_2 shows a decrease near the N $\rightarrow$ A transition point. This has not been observed in other nematics, and might be connected to the rapid increase in α_3. For the (rather few) nematics where α_2 has been measured the ratio $|\alpha_3/\alpha_2|$ is small, of the order of 10^{-2}. As can be seen in Figure 7, this ratio is large in the main part of the nematic interval for 8CB, and increases still

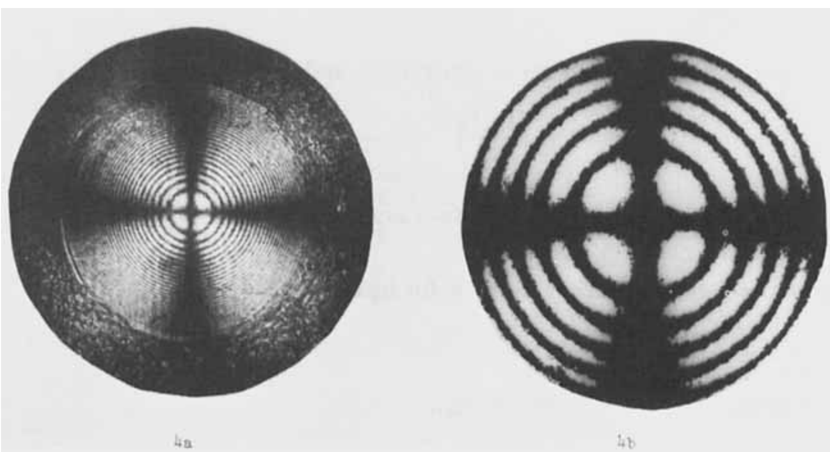


FIGURE 4 a) Appearance of the liquid crystal cell after 90 minutes of torsional shear. $T = 34.5$°C. The boundary between the "ordered" region where $\theta < 90°$ and the outer region with a non-stationary director is seen. b) With an applied electric field the director is stabilized, and a stationary interference pattern is seen.

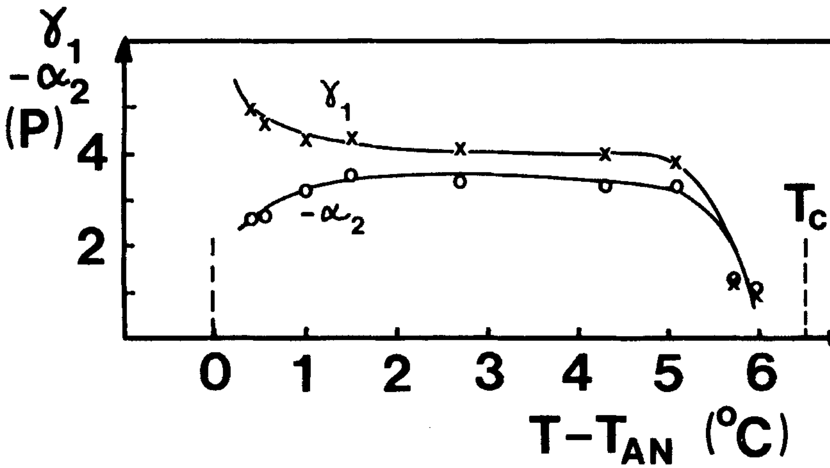


FIGURE 5 Viscosities α_2 and γ_1 as a function of temperature (P = Poises).

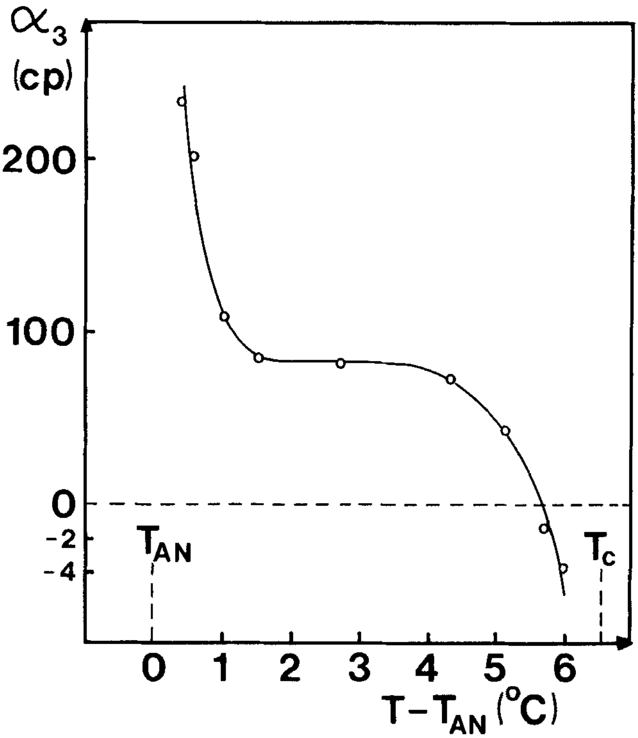


FIGURE 6 Viscosity α_3 as a function of temperature. Note: Enlarged scale for negative values of α_3.

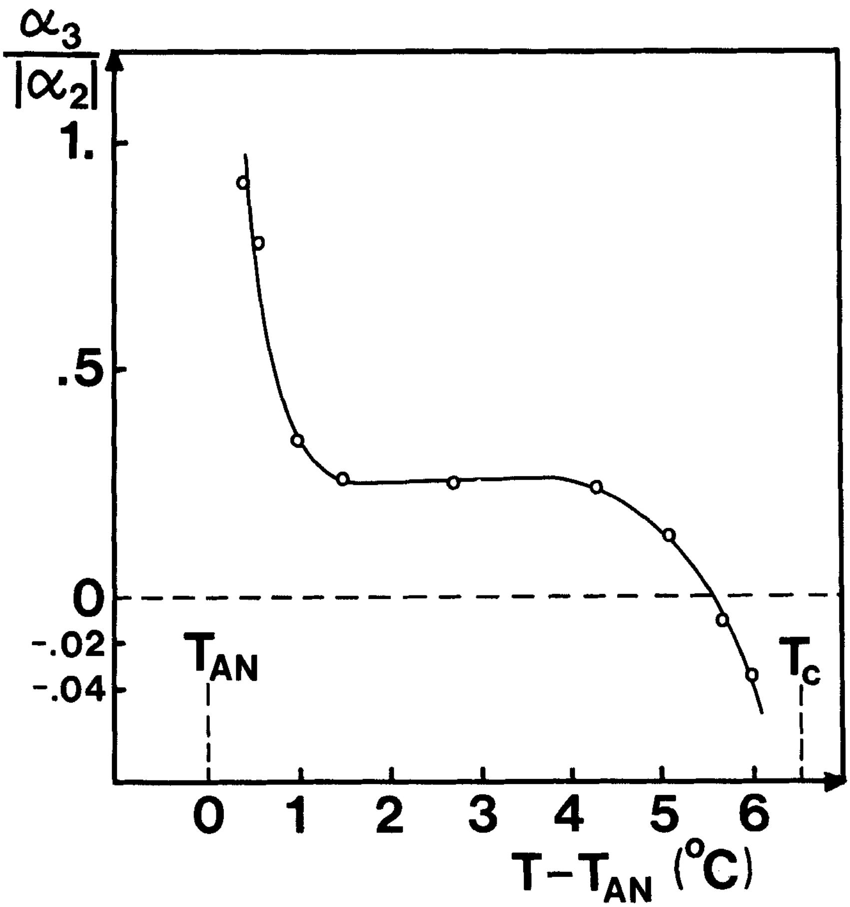
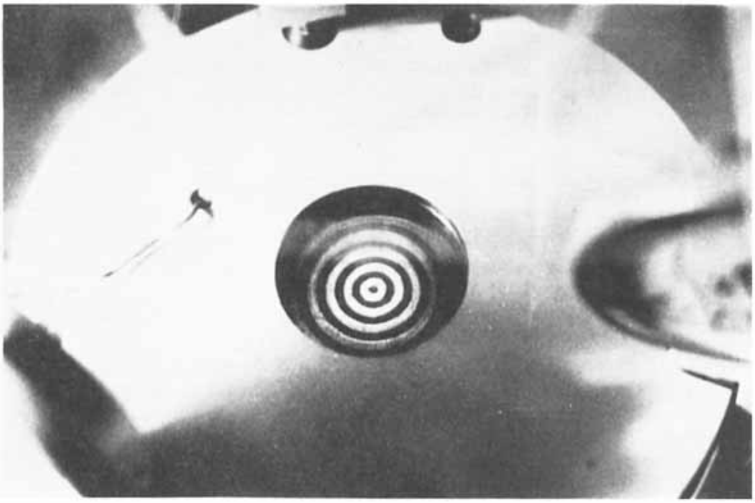


FIGURE 7 The ratio $\alpha_3/|\alpha_2|$ as a function of temperature. Note: Enlarged scale for negative values of the ratio.

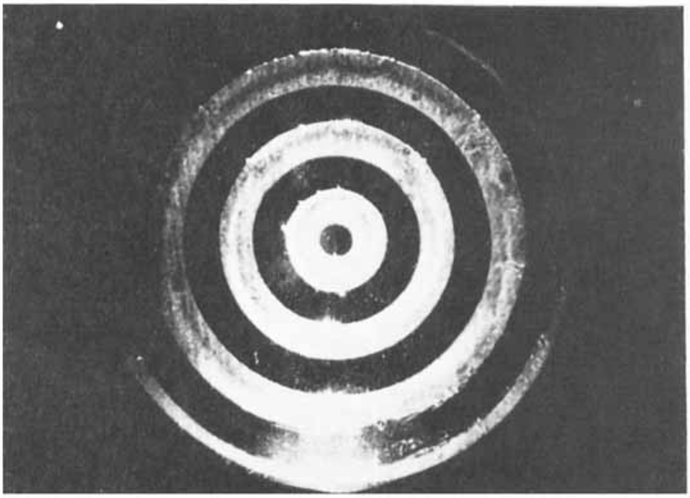
further when the N → A transition is approached. Our experimental curve for 8CB in Figure 7 proves to agree with a proposed qualitative behaviour for CBOOA,[12] although actual measurements of α_3/α_2 has not been reported for CBOOA.

If we look at the situation near the clearing point T_c, we see that α_3 changes sign at $T - T_{AN} = 5.6$°C. This is in good agreement with our earlier experiments,[6] in which the director response to a linear shear was directly observed by conoscopy.

Finally, we show the interesting new structures[6] seen in the torsional shear flow as a result of a large positive α_3. In Figure 8a is seen part of the



8a



8b

FIGURE 8 a) New structures seen in torsional shear when $\alpha_3 > 0$ and without a stabilizing field. b) Close-up of (a) in actual size. Appearance of regular pattern of alternating bands in the liquid crystal. (Dark areas stable homeotropic director; scattering areas unstable, disordered director.)

experimental apparatus, and in the liquid crystal cell at the center bright rings, shown also in Figure 8b. We see here the effect of the shear on the director when a stabilizing electric field is not present.[6] A study of these last mentioned phenomena is in progress.

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